Radiochemical analysis of radionuclides difficult to measure for decommissioning of nuclear facilities

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Abstract:

In decommissioning of a nuclear facility, the radioactivity of various radionuclides has to be measured for estimation of the total inventory of radioactivity and its variation with time. Due to large volume, graphite, concrete, steel and lead are the main low-medium radioactive waste. The radioactivity in these materials comes from many nuclides such as ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{60}Co, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{154}Eu, and some transuranics, in which, the determination of gamma emitters is much easier. But the beta and alpha emitters have to be separated from matrix and other radionuclides before measurement. Most of the beta activity is contributed from ^{14}C, ^{3}H, ^{55}Fe, ^{63}Ni, and ^{41}Ca. Due to high mobility and long half-life of ^{36}Cl, ^{129}I and ^{99}Tc, and high radiation toxicity of transuranics, the determination of these radionuclides in radioactive waste is also important for decommissioning and disposal of the waste. ^{14}C, ^{3}H, ^{36}Cl, ^{63}Ni, ^{99}Tc and ^{129}I are pure beta emitter; except ^{90}Sr, others are low energy beta emitters. ^{41}Ca and ^{55}Fe decay by electron capture. While, most of transuranics are alpha emitters. It is therefore necessary to decompose sample and separate individual radionuclide from matrix element and other interfering nuclides before measurement of their radioactivity. In our laboratory, analytical methods have been developed for the determination of these radionuclides in various nuclear wastes for the decommissioning of nuclear facilities; they are briefly presented.

An oxidizing combustion method has been developed to decompose graphite and concrete, and ^{14}C and ^{3}H released from the sample are measured by liquid scintillation counter (LSC). By this method the sample preparation time can be shortened to only 2-3 minutes. The detection limit of this method for ^{14}C and tritium are 0.96 and 0.58 Bq/g graphite and 0.11 and 0.06 Bq/g concrete respectively. The interference of other radionuclides in samples is insignificant.

A radiochemical separation procedure based on precipitation, ion exchange and extraction chromatography have been developed for the determination of ^{55}Fe and ^{63}Ni. The decontamination factors of the developed method are higher than 10^{4} for the interfering radionuclides. The chemical recoveries for both Fe and Ni are higher than 80%. The detection limits of this method are 0.018 Bq for ^{55}Fe and 0.014 Bq for ^{63}Ni.

Due to high concentration of Ca in concrete, the radioactivity concentration of ^{41}Ca in reactor concrete is relatively high. An alkali fusion method is used to decompose the samples, and various

precipitation methods were developed for the separation of Ca from matrix and other radionuclides, especially for ^{133}Ba, and ^{90}Sr. The purified ^{41}Ca is measured by LSC via counting Auger electrons. The developed method can be used for the simultaneous determination of ^{41}Ca and ^{90}Sr in heavy concrete samples.

The analytical methods for the determination of ^{36}Cl, ^{129}I, and ^{99}Tc were investigated for the analysis of graphite, concrete and steel samples. These radionuclides were first released from samples by acid digestion and combustion, and then separated from matrix and other radionuclides using distillation, ion exchange chromatography and precipitation. The purified radionuclides were measured by LSC.

The methods for the determination of isotopes of Pu, ^{241}Am and ^{237}Np were also developed based on chemical separation using precipitation, extraction and ion exchange and extraction chromatography, the separated transuranics were measured by alpha spectrometry, except for ^{241}Pu by LSC.

The developed method has been successfully applied for the analysis of heavy concrete, graphite, steel, aluminium, and lead from nuclear reactor and the estimation of inventories of these radionuclides in nuclear waste have been carried out. In addition, the methods were also applied for the analysis of sediment, water and vegetation samples for and investigation of the environmental chemical behaviour of these radionuclides.